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Optimizing cellulose fibrillation for the production of cellulose nanofibrils by a disk grinder

Abstract: The fibrillation of a bleached kraft eucalyptus pulp was investigated by means of a laboratory-scale disk grinder for the production of cellulose nanofibrils (CNF), while the parameters disk rotating speed, solid loading, and fibrillation duration were varied. The cumulative energy consumption was monitored during fibrillation. The degree of polymerization (DP) and water retention value (WRV) of the resultant cellulose fibrils were determined as measures of the degree of fibrillation, which was also visualized by scanning electron microscopy, field emission-scanning electron microscopy, and transmission electron microscopy imaging. A higher rotating speed than 1500 rpm did not improve the fibrillation judged by DP and WRV measurements. Solid loading has an insignificant effect on fibrillation in a wide range. The energy consumption (E) was determined as a function of the DP and WRV. The optimal grinding conditions were between 1200 and 1500 rpm at 2.0%–2.2% solid loading.

Keywords: cellulose nanofibrils (CNF), disk grinding, fibrillation efficiency, field emission-scanning electron microscopy (FE-SEM), microfibrillated cellulose (MFC), nanocellulose

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Introduction

Cellulose is naturally present in lignocellulosic biomass as part of the biocomposite consisting of cellulose,

hemicelluloses, and lignin, and the composite has a hierarchical supramolecular structure (Wegner and Jones 2009). Almost half of the plant biomass is cellulose, the basic elements of which are anhydroglucose (AHG) chains. Cellulose is the most abundant biopolymer on earth. Several AHG chains form the elemental fibrils that have very high aspect ratios, specific tensile strength, low thermal expansion, and unique optical properties. From these cellulose subunits, cellulose nanofibrils (CNFs) can be produced. CNF is a broad term to describe many different versions of extremely small-scale fibrillated or crystalline cellulose. CNF is recyclable, renewable, and biodegradable and has a high utilization potential for the production of barrier films, coatings, papermaking additives, paint rheology modifiers, and automotive components, just to mention a few (Siró and Plackett 2010; Klemm et al. 2011; Laka et al. 2011; Dufresne 2012; Freire et al. 2013; Maloney 2015; Žepič et al. 2014).

The mechanical fibrillation of wood fibers is the most common approach for the production of CNFs. A high-pressure homogenizer (Herrick et al. 1983; Turbak et al. 1983; Nakagaito and Yano 2004; Stelte and Sanadi 2009), a microfluidizer (Spence et al. 2011; Zhu et al. 2011), and a disk grinder (Iwamoto et al. 2007; Wang et al. 2012) were used for this purpose. Disk grinding has a high potential for commercial scale-up, as this technology is common in commercial mechanical pulp production. The typical operation conditions are 1.0%–2.0% solid loading with a stone disk rotating at 1500 rpm (Iwamoto et al. 2007; Wang et al. 2012). However, disk grinding for CNF production consumes a lot of high mechanical energy. Our previous study indicated that the energy input for CNF is in the range of 5–30 kWh kg⁻¹ depending on the degree of fibrillation (Wang et al. 2012). Cellulase or chemical pretreatment can reduce the energy input for mechanical fibrillation (Henriksson et al. 2007; Zhu et al. 2011); however, quantitative information is not available. Energy saving is estimated to be 50% based on our laboratory study (Wang et al. 2015).

Disk rotational speed and solid loading are two important parameters affecting energy input in addition to the properties of the feedstock. However, there is no information available concerning the effects of grinding operational parameters for CNF production. These operational

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parameters are grinder specific, and the information about them is proprietary. Furthermore, there is a lack of reliable and quick measures of the degree of fibrillation through mechanical fibrillation. Although transmission electron microscopy (TEM) imaging of CNF can provide excellent visual evidence of the morphology and size (Wang et al. 2012), it is difficult to quantify and evaluate the visual data mathematically. On the contrary, CNF production efficiency can only be evaluated with knowledge of the fibrillation degree and the energy input.

The objective of this study was to explore the possibility of the quantitative characterization of the degree of CNF fibrillation based on the data of the degree of polymerization (DP) and water retention value (WRV) of CNF suspensions. These data along with mechanical energy input should be used for the optimization of CNF production by means of a laboratory-scale disk grinder. The degree of fibrillation should be visualized by scanning electron microscopy (SEM), field emission-SEM (FE-SEM), and TEM imaging. The main motivation of this study is to generate data available for the public as opposed to the proprietary information of grinding operations, which are not accessible.

Materials and methods

Dry and bleached eucalyptus lap kraft pulp (BEP) was obtained from Aracruz Cellulose Brazil. The chemical composition of the pulp was already described by Wang et al. (2012): 78.1±1.0% glucan, 15.3±0.6% xylan, and 0.7±0.1% Klason lignin. BEP (5 kg) was immersed in distilled water overnight at 50% consistency and then re-pulped for 30 min at 2910 rpm in a 50 l laboratory pulper (Voith GmbH, Heidenheim, Germany).

CNF was produced by means of a SuperMassColloider (model: MKZA 6-2, disk model: MDGA 6-80#, Masuko Sangyo Co., Ltd., Japan). The rotation speed (1500 rpm) and consistency (2.0%) from the literature were used as reference (Iwamoto et al. 2007; Stelte and Sanadi 2009; Wang et al. 2012). The present study focused on the rotating speeds of 900, 1200, 1500, and 1800 rpm and solid loadings of 1.8%, 2.0%, 2.2%, and 2.4%. Higher concentrations prevented the flow in the disk chamber due to gelation after 2 h fibrillation and were not investigated. The SuperMassColloider works with two stone disks and is equipped with an energy meter to record energy input. The bottom disk is rotating, whereas the upper disk is stationary. Initially, the disk gap was dynamically set to zero (two disks just touch each other) without pulp. Then, with the addition of pulp, the gap was adjusted down to 100 µm. The gap was kept constant for all experiments. Due to the presence of pulp suspension or slurry, there was no direct contact between the two grinding disks even with a negative clearance. Pulp suspension was fed into the hopper continuously through a plastic hose, which was driven by a peristaltic pump (Cole Parmer, Chicago, IL, USA). Pulp suspension was fed under gravity from the hopper to the disk chamber. The fibrillated pulp suspension was discharged by centrifugal force, and the time-dependent energy consumption was recorded at 30-min intervals.

The DP of fibrillated cellulose is a measure of the degree of fibrillation (Shinoda et al. 2012; Wang et al. 2012). The DP of CNFs was measured according to TAPPI Standard Test Method T230 om-99 (TAPPI 2009). CNF (0.2 g; based on oven dry weight) was first dissolved in 40 ml of 0.25 M cupriethylenediamine solution, and the viscosity of the solution was determined in a capillary viscosimeter (duplicate experiments). The DP was calculated according to Mazumder et al. (2000):

$$DP^{0.905} = 0.75 [954 \log(X) - 325] \quad (1)$$

where X is the measured viscosity.

The WRV was measured according to a modified Scandinavian test method SCAN-C 62:00. The CNF suspension was diluted to approximately 2.0% solid consistency. A 10 g CNF suspension was wrapped in a tarred 10 µm nylon screen filter and placed in a centrifuge tube with a lower metal mesh support to leave a space for water accumulation at the bottom of the tube. The CNF sample was centrifuged at 3000 g for 15 min (International Centrifuge EXD 4871, International Equipment Co., Chattanooga, TN, USA). The CNF sample was weighed before and after oven drying overnight at 103±2°C. Again, duplicate experiments were carried out.

$$WRV (\%) = 100 (m_{\text{wet}} - m_{\text{od}}) / m_{\text{od}} \quad (2)$$

where m_{wet} and m_{od} are the mass of the centrifuged CNF sample before and after oven drying, respectively.

The specimens for SEM were prepared by drying drops of the aqueous slurry of 0.2 g l⁻¹ on aluminum mounts. All SEM specimens were sputter coated with gold to provide adequate conductivity for examination in a Zeiss EVO 40 SEM (Carl Zeiss NTS, Peabody, MA, USA) under ultrahigh vacuum conditions. The FE-SEM images were obtained by an FE-SEM (Hitachi SU-70) together with Schottky-type field emission gun at the University of New Brunswick (Fredericton, New Brunswick, Canada). The acceleration voltage of 3.0 kV and a beam current of a few nanoamperes were employed to reduce surface charging, which prevented the beam damage effects on the sample and led to high-resolution images.

For TEM, aqueous CNF (0.2 g l⁻¹) was further diluted in water and sonicated to disperse the particles. The TEM grids (ultrathin carbon films supported by fenestrated carbon films) were floated on drops of approximately 5 µl sample for 1–2 min and were then swished through two consecutive 250 µl drops of 2% aqueous uranyl acetate. Excess stain was removed by capillary action and gentle blotting resulting in negatively stained particles. The samples were imaged by a Philips CM100 TEM (FEI Co., Portland, OR, USA) with an accelerating potential of 100 keV. The images were captured on Kodak SO-163 electron image film and later scanned into digital images at 600 dpi resolution.

Results and discussion

Energy consumption

The cumulative energy inputs (without load) for fibrillation increased linearly with increasing fibrillation time at different rotating speeds (Figure 1a), in agreement with the data of Wang et al. (2012). The cumulative energy also

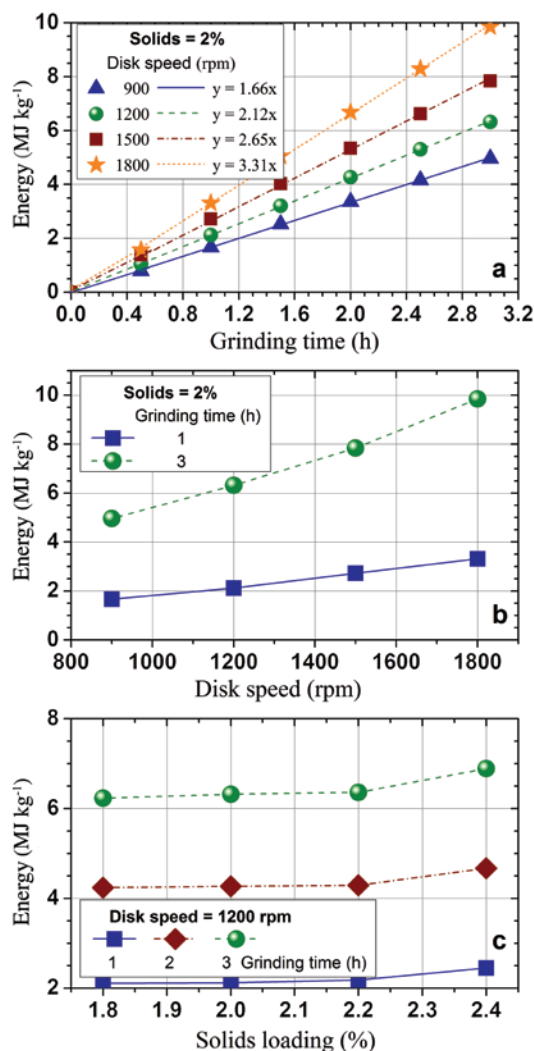


Figure 1 Effects of various operating parameters on the energy consumption of disk grinding for the fibrillation of bleached kraft eucalyptus pulp: (a) grinding time, (b) disk speed, and (c) solid loading.

increased almost linearly with disk speed at a given solid loading of 2.0% (Figure 1b), demonstrating that the rotating speed is a major factor controlling the time-dependent energy consumption. Based on this observation, it is obvious that energy consumption is proportional to the effective amount of material being fed into the grinder. High rotating speed increases the cellulosic material feeding rate and results in an increased effective (total) amount of fibrillated material and high energy consumption. The slope of energy consumption with respect to rotating speed increased at longer times (Figure 1b) due to the cumulative effect of increased rotating speed. Solid loading had a negligible effect on energy consumption below 2.2% loading (Figure 1c). However, increased solid loading can increase shearing and rolling friction

resistance inside the disk chamber, resulting in higher energy consumption for fibrillation as evidenced at solid loading of 2.4% (Figure 1c).

Degree of polymerization

The DP of fibrillated cellulose decreased continuously with grinding time (Figure 2a). The DP was reduced from approximately 900 to 660 and 600 after 3 h grinding of 2.0% solid consistency at 1800 and 1500 rpm, respectively. At 2.0% solids, maximum DP reduction occurred at 1500 rpm, with lesser effects at speeds higher or lower than this. With a disk speed of 1500 rpm, a minimum DP of approximately 600 was achieved after 3 h grinding

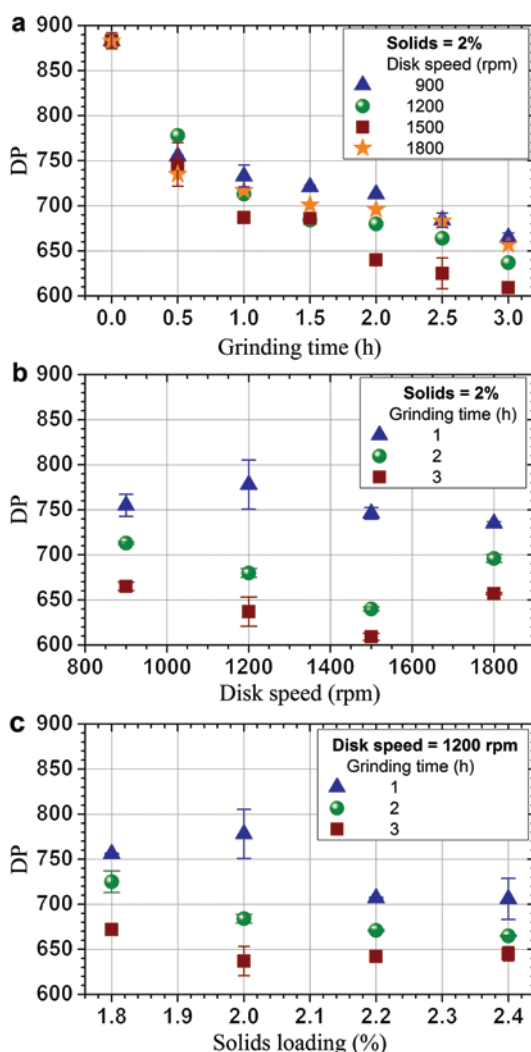


Figure 2 Effects of various operating parameters on the DP of the resultant cellulose fibrils: (a) grinding time, (b) disk speed, and (c) solid loading.

(Figure 2a). However, with a further increase in disk speed to 1800 rpm, the DP was higher than that achieved at 1500 rpm after more than 1 h of grinding (Figure 2b). The higher disk rotating speed of 1800 rpm did not improve cellulose fibrillation regardless of the higher energy input (Figure 1a and b). The higher the rotating speed is, the higher is the feeding rate and the shorter is the resident time in the disk chamber for nanofibrillation. However, more cycles did not improve nanofibrillation performance because a high speed might have created slips among fibrils to result in reduced frictions for fibrillation. The resident time of the pulp suspension or slurry inside the grinding chamber, which was mainly dictated by the rotating speed, is an important parameter that affects the overall nanofibrillating performance (Figure 2b). The optimal rotating speed was 1500 rpm, which produced cellulose fibrils with a minimum DP of approximately 600.

The residence time was affected by the viscosity of the pulp slurry that is, in turn, influenced by solid loading and temperature. The temperature of the pulp slurry was controlled by the cooling of the discharged pulp slurry. The solid loading in the range of 1.8%–2.4% on DP reduction was negligible if the grinding time was longer than 1 h (Figure 2c). This was partially due to the gelation of the fibrillated pulp at solid consistency higher than 2.4%.

Determination of cellulose fibrillation efficiency

The derivative of the DP with respect to grinding energy, $d(DP)/dE$, under a set of conditions, such as disk speed and solid loading, was evaluated as a measure of cellulose fibrillation efficiency. This approach takes also the grinding energy cost into account, and it is a better measure than the simple DP alone. Four data sets with this regard are plotted in Figure 3. Linear regression analysis was carried out to obtain the slopes for $d(DP)/dE$ plots. The effect of disk speed at 2% solid loading on $d(DP)/dE$ was examined as shown in Figure 4a. The error bars are fitting errors and represent 95% confidence interval. The results suggest that the grinding efficiencies are approximately the same for disk speeds between 900 and 1500 rpm, as they are all within the fitting errors. However, $d(DP)/dE$ at 1800 rpm was significantly lower (in absolute value) than those at the three lower disk speeds; that is, it was reduced from -23.3 ± 4.1 at 1200 rpm to only -8.6 ± 0.8 at 1800 rpm, which was a 60% reduction. This finding is substantial, as it is significantly larger than the fitting errors (Figure 4a). Thus, a moderate disk speed of 1200 rpm is more energy efficient and may be preferred.

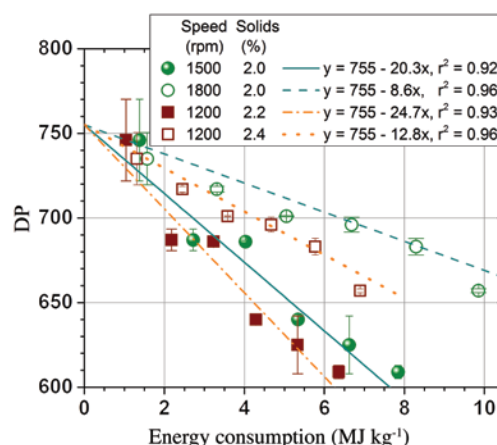


Figure 3 Correlations between the DP of fibrillated cellulose fibrils and grinding energy consumptions.

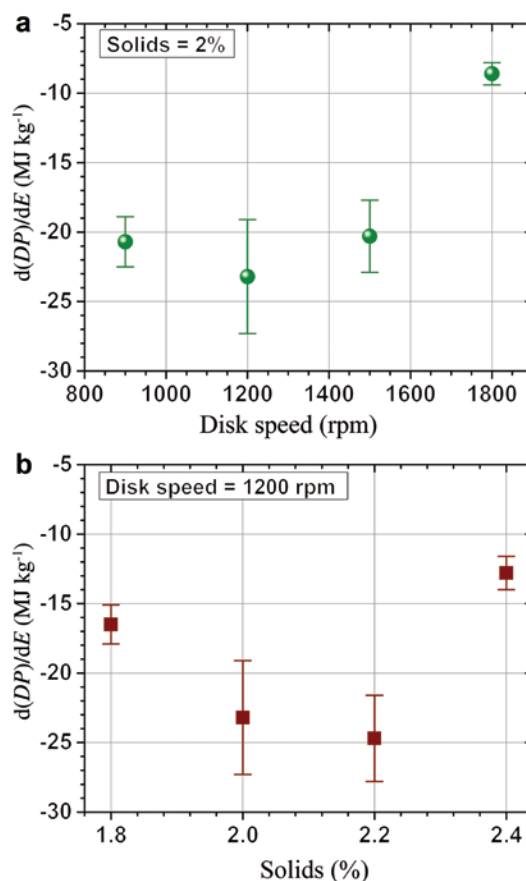


Figure 4 Effects of grinding conditions on the fibrillation efficiency measured by $d(DP)/dE$: (a) disk speed and (b) solid loading.

The solid loading increment from 1.8 to approximately 2% increased $|d(DP)/dE|$ (Figure 4b) from 16.5 ± 1.4 to 23.2 ± 4.1 . This is probably due to the increased friction or shear actions between cellulose fibrils, which facilitate

fibrillation and DP reduction. However, a further increase in solid loading of approximately 2.4% significantly reduced $|d(DP)/dE|$ from 24.7 ± 3.1 at 2.2% to 12.8 ± 1.2 at 2.4%, which was a substantial 50% reduction based on the fitting errors (Figure 4b). High solids may significantly increase the friction forces among fibrils but, under these conditions, less fibril cutting occurs. Thus, processing with high solid content is preferable. Based on the data in Figure 4a and b, the most efficient processing conditions are between 1200 and 1500 rpm at 2.0%–2.2% solid loading.

Water retention value

The WRV was tested to gauge the effects of heating, drying, and delignification on water-accessible surface areas of fibers and lignocelluloses (Welf et al. 2005; Luo and Zhu 2011; Williams and Hodge 2013). Fibrillation through disk grinding causes the delamination and defibrillation of fiber cell wall, and these events entail an increment of fibril surface area accessible to water. Therefore, the WRV is a measure of CNF surface and swelling ability. Also in the present study, the WRV increased continuously with grinding time under different rotating speeds (Figure 5a) and different solid loadings (Figure 5b). The variations of WRV with grinding speed and solid loading had similar trends as those of the DP. The WRV increased with disk speed up to 1500 rpm and reduced significantly at a disk speed of 1800 rpm, in agreement with increased DP at a disk speed of 1800 rpm for grinding times longer than 2 h (Figure 2b). The effects of solid loading on WRV were not significant in the range of 1.8%–2.4%, similar to the observation in the context of DP measurements (Figure 2c). The gelation of the pulp slurry prevents solid loadings >2.4%. Fibrillation had to be stopped at 2–2.5 h in the case of solid loading of approximately 2.5% because the flow was inhibited.

The slopes of the linear regression lines of $d(WRV)/dE$ are listed in Table 1, which are in agreement with those presented in Figure 4a and b. These data also suggest that the optimal grinding conditions for the grinder applied were 1200–1500 rpm at 2.0%–2.2% solid loading, as was also indicated by DP measurements.

Electron microscopy

According to SEM and TEM imaging, the prerefined BEP fibers have typical diameters of approximately 20 μm with a length of 1 mm (Figure 6a). Under combined

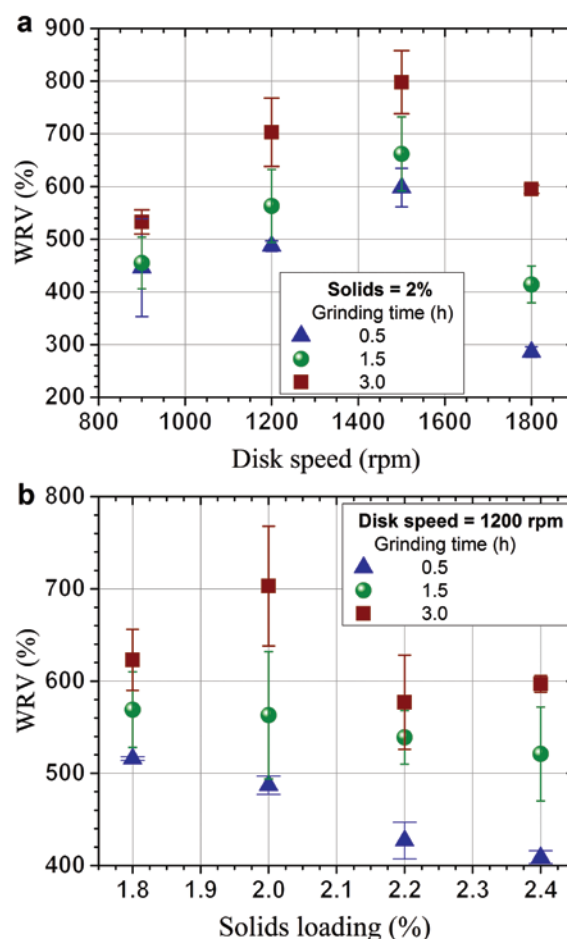


Figure 5 Effects of grinding conditions on the WRV of the resultant cellulose fibrils: (a) disk speed and (b) solid loading.

Table 1 Fibrillation efficiency evaluations based on the derivative $d(WRV)/dE$, where WRV is water retention value and E is grinding energy ($\pm$ SD from duplicate experiments).

Disk speed (rpm), solid content (%)	WRV at 3 h	$d(WRV)/dE$
At 2% solid		
900 rpm	533 $\pm$ 23	19.2 $\pm$ 6.5
1200 rpm	703 $\pm$ 65	40.2 $\pm$ 2.0
1500 rpm	798 $\pm$ 60	30.6 $\pm$ 2.5
1800 rpm	595 $\pm$ 8	37.3 $\pm$ 0.1
At 1200 rpm		
1.8%	623 $\pm$ 33	20.9 $\pm$ 1.1
2.0%	703 $\pm$ 65	40.2 $\pm$ 2.0
2.2%	577 $\pm$ 51	23.4 $\pm$ 5.3
2.4%	597 $\pm$ 9	33.7 $\pm$ 1.4

compressive, shearing, and friction forces, both internal and external fibrillation took place (Hartman 1985; Kerekes 2005; Wang et al. 2012). After 1.5 h of grinding (Figure 6b), the fibrils were separated and partly

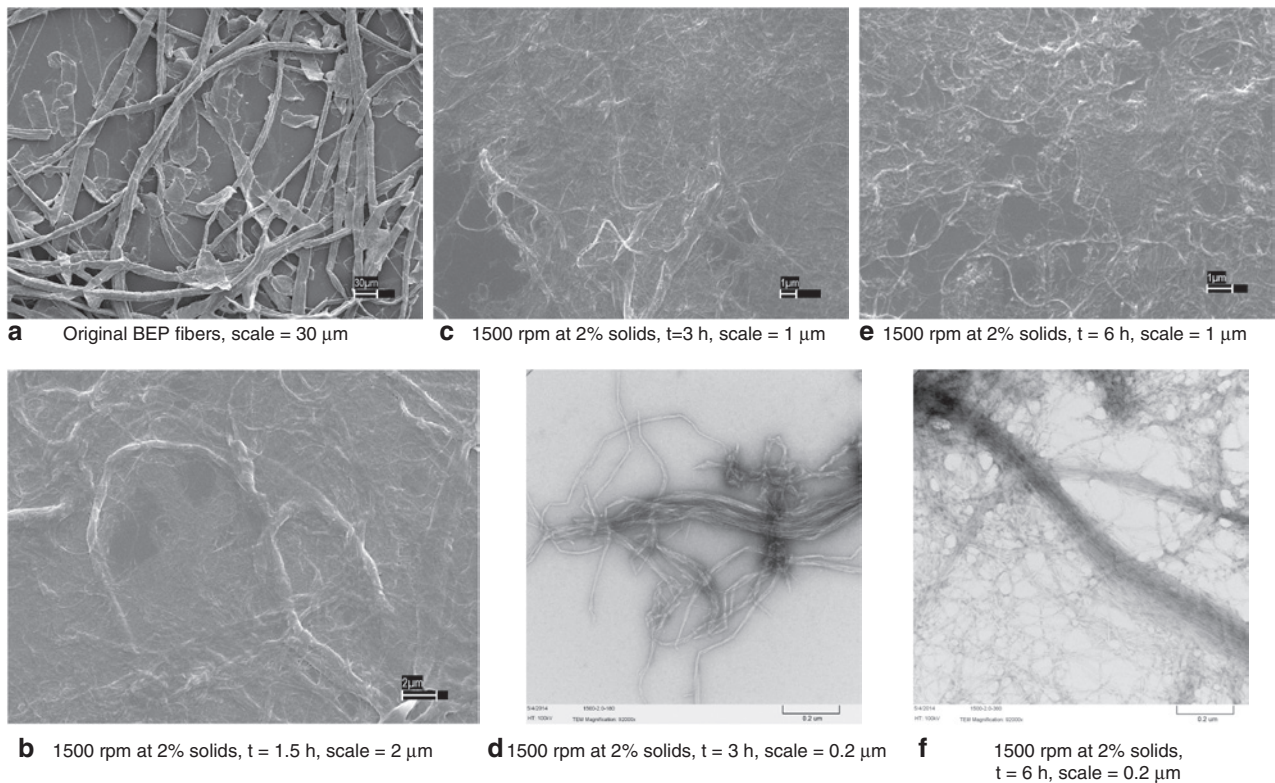


Figure 6 SEM and TEM images showing the progression of fibril morphology with grinding time at a grinding speed of 1500 rpm with 2% solid content: (a) SEM: original bleached eucalyptus fibers (scale, 30 μm), (b) SEM: 1.5 h (scale, 2 μm), (c) SEM: 3 h (scale, 1 μm), (d) TEM: 3 h (scale, 0.2 μm), (e) SEM: 6 h (scale, 1 μm), and (f) TEM: 6 h (scale, 0.2 μm).

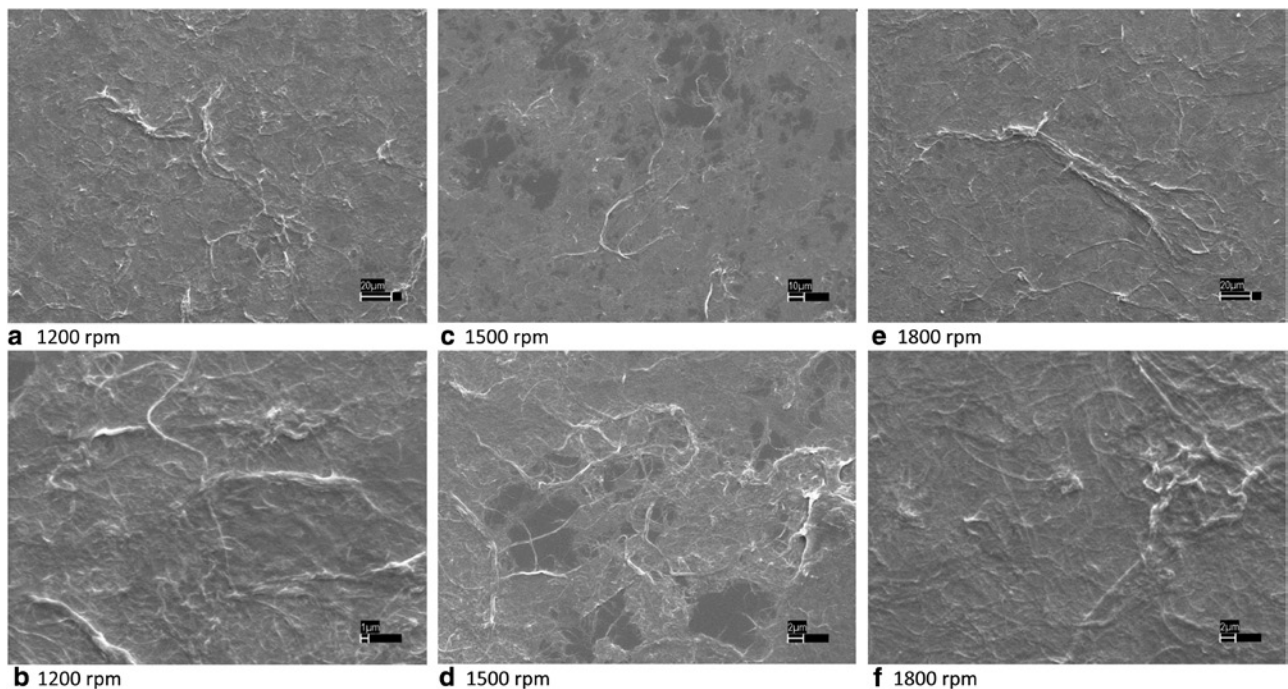


Figure 7 Comparisons of fibril morphologies by SEM at three grinding speeds with constant grinding time of 3 h and 2% solid content: (a) 1200 rpm (scale, 20 μm), (b) 1200 rpm (scale, 1 μm), (c) 1500 rpm (scale, 10 μm), (d) 1500 rpm (scale, 2 μm), (e) 1800 rpm (scale, 20 μm), and (f) 1800 rpm (scale, 2 μm).

cut and the average fibril diameter was approximately 1 μm . Processing to 3 h lowered the diameters below 1 μm (Figure 6c). TEM images illustrate the ultrastructure of the fibrils with diameters between 20 and 100 nm (Figure 6d). Figure 6e to d (SEM and TEM after 6 h) shows the changes in comparison to 3 h grinding (Figure 3c). The TEM image after 6 h grinding (Figure 6f) illustrates the presence of fibrils at the nanoscale level, where the fibrils were completely entangled. Kinks on fibrils are visible (TEM in Figure 6d). Wang et al. (2012) also made similar observations.

Both the DP (Figure 2a) and the WRV (Figure 5a) measurements indicate that a disk grinding speed of 1500 rpm produced finer fibrils than 1200 and 1800 rpm. The SEM images verify these macroscale characterizations, whereas Figure 7a–f (3 h grinding at 2% solid load) shows that a grinding speed of 1500 rpm (Figure 7c and d) produced finer fibrils than either 1200 rpm (Figure 7a and b) or 1800 rpm (Figure 7e and f). Most of the fibrils were <1 μm with 1500 rpm (Figure 7d), whereas many fibrils were in the order of 2 μm with 1200 rpm (Figure 7b) and 1800 rpm (Figure 7f). Fibrils with diameters as large as 10 μm were also seen with 1200 rpm (Figure 7a) and 1800 rpm (Figure 7e). The derivatives of the DP and WRV with respect to grinding energy are good measures for the estimation of the degree of nanofibrillation.

Conclusions

The DP and WRV of the resultant cellulose fibrils are good measures for the determination of the degree of fibrillation. TEM imaging revealed that fibril morphologies were consistent with those measured by the DP and WRV. The fibrillation efficiency was also determined based on the derivatives of the DP or WRV with respect to the fibrillation energy (E). A high rotating speed of 1800 rpm was not efficient and did not improve the performance of fibrillation as measured by both the DP and WRV. The optimum grinding conditions were between 1200 and 1500 rpm at 2.0%–2.2% solid loadings, which can result in fibrils with an average DP and WRV of approximately 600 and 750%, respectively.

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